On Two Models of the Light Pulse Delay in a Saturable Absorber

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Abstract

A comparative analysis of two approaches to description of the light modulation pulse delay in a saturable absorber is presented. According to the simplest model, the delay of the optical pulse is a result of distortion of its shape due to absorption self-modulation in the nonlinear medium. The second model of the effect, proposed at the beginning of our century, connects the pulse delay with the so-called "slow light" resulting from the group velocity reduction under conditions of the coherent population oscillations. It is shown that all the known experimental data on the light pulse delay in saturable absorbers can be comprehensively described in the framework of the simplest model of saturable absorber and do not require invoking the effect of coherent population oscillations with spectral hole-burning and anomalous modifications of the light group velocity. It is concluded that the effect of group velocity reduction under conditions of coherent population oscillations has not received so far any experimental confirmation, and the assertions about real observation of the "slow light" based on this mechanism are groundless.

INTRODUCTION

Saturable absorption is a basic effect of the incoherent nonlinear optics. In its simplest form, the effect is revealed as bleaching of the medium with increasing intensity of the light passing through it. This effect became known still in the pre-laser epoch due to experimental works of Kasler's group on optical orientation of atoms [1]. Its observation under relatively low light power densities was possible because of low population relaxation rates in these systems. Later on, after the advent of laser, this effect has been studied in great detail [2, 3, 4] and found application in the laser systems of Q-switching and mode-locking (see, e.g., [5, 6]). Nonlinearity of the saturable absorber is determined not by nonlinearity of polarizability of the medium at optical frequencies, but rather by the dependence of optical constants of the medium (optical absorption) on the light intensity or on the light power density. One can say that the carrier frequency of the light field and, moreover, even the fact of its existence (see, e.g. [7]) play no role in this effect. Under consideration is, in essence, the dynamics of penetration of the energy flux through a barrier whose transmissivity depends on this flux.

Mechanism of the effect is usually determined by the light-induced changes in populations of the states of the system and, for this reason, the response of the absorber exhibits a certain sluggishness associated with the sluggishness of the population relaxation. As a result, the light pulse transmitted through the saturable absorber appears to be distorted or, under certain conditions, shifted in time. The sign of this shift, in the general case, can be both positive and negative. All the regularities of interaction of light with a saturable absorber have been studied in great detail to early 70's [2, 3, 4] and, later on, the interest to these effects was, to a considerable extent, lost.

At the beginning of our century, however, the effects of retarded response of saturable absorber have been rediscovered and reinterpreted [8, 9]. In the new interpretation, the pulse delay was associated with a low group velocity of light propagating in the *linear medium* with a highly steep refractive

index dispersion. The high steepness of the dispersion, in authors' opinion, was provided by the narrow spectral hole burnt by laser light in the homogeneously broadened absorption band under conditions of the coherent population oscillations (CPO) [10]. In this interpretation, small temporal shifts of the light pulse passed through the medium ('small' in the scale of the pulse length, but huge in the scale of time needed to the light to pass through the sample with the velocity c) were converted into giant factors of the group velocity reduction. With appearance of the novel model of pulse delay in a saturable absorber, the slow light technique, which had in its arsenal, at that time, only the method of electromagnetically induced transparency, was enriched by one more experimental approach, which allowed one to demonstrate achievements of the same (or even higher) level under much simpler experimental conditions at room temperatures on solid-state objects, highly convenient for practical applications (see, e.g., [11, 12]).

Thus, it occurred so that one and the same phenomenon was described in the framework of two, fundamentally different and incompatible models. The discussion aroused on this issue in literature [13, 14, 15, 16, 17, 18, 19, 20, 21] has not lead to a consensus.

In this paper, we present a comparative analysis of the two models of the light pulse delay in a saturable absorber and show that the effect of the CPO-based reduction of the light group velocity has not been reliably observed so far, all the relevant experimental data on pulse delay perfectly agree with the simplest model of saturable absorber, and there are no grounds to consider the CPO-based slow light as really existing.

PULSE DELAY AS A RESULT OF ITS SHAPE DISTORTION IN A SATURABLE ABSORBER

Most essential properties of a saturable absorber can be treated neglecting the propagation effects (for more detail, see [14]). For that, it suffices, in the

expression describing in the general form the nonlinear relation between the intensity of the light transmitted through the absorber (I_{out}) and intensity of the incident light (I_{in})

$$I_{out} = K(I_{in}, t)I_{in}, \tag{1}$$

to take into account, in the simplest form, the aforementioned sluggishness of the transmission dynamics of the medium

$$\frac{dK}{dt} = \frac{K_{eq} - K}{\tau} \tag{2}$$

(K_{eq} is the equilibrium value of the transmissivity K for the current intensity I_{in}), and to restrict oneself to the linear term of expansion of the transmissivity K_{eq} (implying relatively small variations of the intensity I_{in})

$$K_{eq}(I_{in}) = K_0 + K_1 I_{in}, K_1 I_{in} \ll K_0, \tag{3}$$

Relations (1)–(3), in spite of the used simplifications, make it possible to describe all the main regularities of response of such a nonlinear medium to intensity variations of the acting light. In particular, the frequency dependence of the intensity modulation amplitude I_ω exhibits a Lorentzian feature (peak or dip, depending on sign of the constant K_1) with a half-width of τ^{-1}, centered at zero frequency (Fig. 1a), while the corresponding dependence of the intensity modulation phase shift ϕ_ω shows the greatest delay (respectively, positive or negative) at the frequency $\omega \sim \tau^{-1}$ (Fig. 1b). The light pulse, in the general case, appears to be distorted at the exit of the medium. However, for a smooth pulse of sufficiently large duration ($> \tau$), this distortion is reduced practically to a pure shift, with its sign being also determined by the sign of the coefficient K_1 (Fig. 1c). In more detail, all these regularities are described in [2, 4, 14].

These are the dependences that were demonstrated in the first publications on the "CPO-based slow light" and that are considered so far as good evidences for the reduction of the light group velocity in saturable absorbers.

The dependences presented in Fig. 1 describe unavoidable properties of a saturable absorber that have nothing to do with anomalous modifications of the light group velocity in the medium. In particular, the above effects of pulse delay or delay of the light intensity modulation can be observed in media of ultimately small thickness in the light of arbitrary spectral composition. The pulse delay is, in essence, a result of light self-modulation, and the pulse shape changes when the light-induced bleaching (or darkening) of the medium occurs in the process of the pulse propagation, and absorption of the medium for the leading and trailing edges of the pulse proves to be different.

It is noteworthy that Eqs. (1)–(3) describe, along with transformation of temporal behavior of the light transmitted through a saturable absorber, transformation of its *intensity spectrum*. The saturable absorber, with respect to the transmitted light, plays the role of the light-controlled optical modulator. However, in view of Eq. (3), high frequencies of the light intensity modulation do not participate in controlling the modulator. For this reason, the main distortions of the intensity spectrum occur in the range of low frequencies ($\omega < \tau^{-1}$). In terms of the conventional spectroscopic technique, the most direct way to make sure that this is the case is to make use of the light with a "white" intensity spectrum (the light with its intensity modulated by a "white noise"). Then, at the exit of the medium, the light intensity spectrum (originally flat) will show a peak (or a dip) centered at zero frequency with the width τ^{-1}. Usually, however, the frequency dependences of this kind are obtained using the light with "monochromatic" intensity spectrum: the light intensity is subjected to a weak harmonic modulation, with the phase and amplitude of the modulation being measured at the exit of the medium as a function of frequency. Thus, the dependences presented in Fig. 1 can be considered as a result of filtration of the light intensity spectrum by a saturable absorber.

PULSE DELAY AS A RESULT OF GROUP VELOCITY REDUCTION

Let us now turn to a more sophisticated model that connects the pulse delay in a saturable absorber with changing group velocity of light. The effect of coherent population oscillations (CPO) lying in the basis of this model exploits the same aforementioned frequency characteristics of the response (Fig. 1a and 1b). Now, however, they are implied to demonstrate changes in *optical spectrum* of the light transmitted through the saturable absorber, rather than changes in its *intensity spectrum*.

The CPO effect is observed as follows. A saturable absorber pumped by a monochromatic light wave (of partially saturating intensity) with the frequency ν_0 is illuminated by a weak (probe) monochromatic wave with a shifted frequency of $\nu_0 + \delta\nu$ (Fig. 2). As a result of beats of the two waves with close frequencies, intensity of the total light field appears to be modulated at the difference frequency $\delta\nu$ (Fig. 2b). We know, however (see Fig. 1a), that the light intensity modulation depth at the exit of the saturable absorber changes when the modulation frequency is comparable with the inverse relaxation time of the absorber. In particular, the bleachable absorber amplifies modulation of the transmitted light (Fig. 2c). As a result, this inevitably affects *optical spectrum* of the field. There arise two symmetric sidebands $\nu_0 \pm \delta\nu$, so that one of them exactly coincides with the probe wave both in phase and in frequency, thus leading to enhancement of the latter at the exit of the absorber (Fig. 2d). As a result of this two-wave interaction, the transmission spectrum of the probe wave in the vicinity of the pump acquires the shape of a peak with the width $\sim \tau^{-1}$, centered at the frequency of the pump ν_0 (Fig. 3a). This is the gist of the CPO effect. The hole in the absorption spectrum of the medium burnt in this way is, at low intensities of the probe wave does not depend on the probe beam intensity, and the saturable absorber in the field of a resonant monochromatic pump can be considered as a linear "supermedium" with a narrow dip in the absorption

spectrum at the frequency of the pump. If this is the case, then such a medium, in conformity with the known Kramers-Kronig relations, should exhibit, in the vicinity of the dip, the region of a highly steep dispersion (Fig. 3b).

Recall that at high steepness of dispersion of the refractive index n ($\omega\frac{dn}{d\omega} \gg 1$), the value of the light group velocity in the medium

$$V_{gr} = \frac{c}{n + \omega(dn/d\omega)} \tag{4}$$

is determined by this *dispersion-related* contribution. Thus, the saturable absorber, in the framework of such a model, allows one to realize situation similar to that used in the first successful experiments on slow light based on the effect of electromagnetically induced transparency. [22, 23]. In this case, the group velocity of the light pulse, with its spectrum lying within the width of the dip, should strongly differ from the phase velocity of light in the medium. In particular, in the bleachable absorber, dispersion of the refractive index in the vicinity of the central frequency of the dip will be *normal* ($\frac{dn}{d\omega} > 0$), and, correspondingly, the group velocity of the "resonant" probe pulse will be anomalously low ("slow light"), while in the reverse absorber with anomalous dispersion in the region of the dip ($\frac{dn}{d\omega} < 0$), group velocity of the light will be anomalously high ("fast light").

This model was used to interpret the experiments carried out with the ruby and alexandrite crystals [8, 9] that were laid into the basis of the new slow light technique.

It should be noted, however, that this model implies another type of the experiment. While in the first case (pulse distortion in a saturable absorber) one studies optical response of the medium to intensity modulation of the light with arbitrary spectrum, the new model deals with the response of the medium pumped by a monochromatic (quasimonochromatic) field to the light pulse with a sufficiently narrow spectrum, resonant with respect to the pump. Therefore, these two models, strictly speaking, describe different ex-

periments. In reality, however, the second model was used to interpret simple single-beam experiments with saturable absorbers. This has eventually lead to collision of the two models.

DISCUSSION

The start of this collision was given by the papers on "hole burning" in homogeneously broadened absorption spectra of the ruby and alexandrite crystals [24, 25], published long before the idea of "slow light" was born.

As was mentioned above, the dip (or the peak) in the modulation spectrum of a saturable absorber at zero frequency is its unavoidable feature that should be necessarily observed. On the other hand, the effect of coherent population oscillations is the manifestation of the same feature, but under essentially different experimental conditions, when the intensity modulation is formed by superposition of two fields with close frequencies (strong and weak), when one of them (weak) is monitored. The dip in the intensity modulation spectrum by itself, reflecting basic properties of the saturable absorber, evidently, cannot serve as an evidence for the dip in the optical spectrum.

Still, the conclusion about spectral hole-burning under conditions of CPO has been made in [24, 25] based solely on observation of the dip in the intensity modulation spectrum. According to the proposed model, the dip in the optical spectrum of the saturable absorber is detected by sidebands of the modulated laser beam and is created by the field of carrier frequency. The width of the dip lied in the range of tens of Hz, and the conclusion made by the authors evidently could not be confirmed by direct spectroscopic measurements. In these papers, the questions about spectral width of the laser source and, therefore, about meeting conditions needed for observation of the CPO effect, were not discussed.

The idea of using the narrow spectral dip arising under conditions of CPO

for obtaining slow light was borrowed from the papers [22, 23], where a similar dip arose under conditions of electromagnetically induced transparency [8, 9]. The amplitude measurements of the modulation spectra [24, 25] were complemented by temporal measurements. The results of these measurements completely agreed with the simplest model of the saturable absorber [2, 4], but, in accordance with papers [24, 25], they were interpreted in terms of the "CPO-based slow light".

The new interpretation of pulse delay in a saturable absorber had indeed brought this effect to the front line of science, while the simplicity of the experiments in combination with the scale of the easily achievable values "group indices" (corresponding to group velocities down to fractions of mm/s [26]) rendered this slow light technique highly popular.

All the research regarding the light pulse delay in saturable absorbers after that was carried out exclusively in the framework of the "slow light" concept, which postulated mechanism of the effect by its title, and as a result, no alternative models of the pulse delay could be considered. Meanwhile, the new model did not look convincing.

The first question arising in the new interpretation of phase delay of the harmonically modulated light was related to spectral width of the laser light. How could the light source with the line width lying in the range of hundreds MHz create the spectral dip with a width of several tens of Hz? Attempts to explain this paradox were made much later in terms of a special model of the laser field [16]. It is appropriate to note here that, in full agreement with regularities of behavior of the saturable absorber, all the indications of the "CPO-based slow and fast light" were observed in incoherent light of thermal sources [27], when the model of the laser field proposed in [16] cannot be valid.

The second problem mentioned already in [8] was that the proposed theory could not explain the delay of a single pulse in the absence of pump. In authors' opinion, this was a unique case when the saturating pulse produced

its own pump field.

It is noteworthy that practically in all studies of the "CPO-based slow light" the experiment was performed using a single beam whose intensity was modulated in a harmonic or pulsed fashion. In other words, the conditions for observation of the CPO effect (that imply separation of the probe beam from the pump) were not satisfied. An exception was the paper [28] that demonstrated experimentally the fact that the hole burnt by a monochromatic pump in the spectrum of a saturable absorber under conditions of CPO is correlated, in conformity with the Kramers-Kronig relations, with a dispersion curve, highly steep at the center of the hole. Direct measurements of the light pulse delay, in this paper, were not performed. An attempt to observe the CPO-based group velocity reduction with separation of the pump and probe beams was made in [29]. However, the object for study (an ensemble of quantum dots) was characterized by a strong inhomogeneous broadening, and, for this reason, the experiment was reduced to detection of spectral hole in the inhomogeneously broadened absorption band with observation of the related slow light as in [30]. In recent years, a considerable number of papers have been published that convincingly showed that all the known experiments on the "CPO-based slow light" can be described, in a more natural way, in the framework of the simplest model of saturable absorber, without invoking the effect of coherent population oscillations, spectral hole burning, and anomalous modifications of the light group velocity [7, 13, 14, 17, 18, 20, 31]. In some papers, there have been described experiments that came into direct conflict with the "slow light" model. In particular, in [20], there has been demonstrated possibility to control the light pulse delay in a saturable absorber using the pump beam strongly shifted with respect to that of the probe light. In [27], there have been demonstrated all the features of the "light with negative group velocity" and "ultraslow light" on simple photochromic objects using an incoherent light source (incandescent lamp), when the condition for the CPO definitely could not be met. Universality of the effects of delay of modulated signals in the nonlinear systems

described by Eqs. (1)–(3) has been shown in [7] using as an example the electric circuit with a nonlinear resistor, where the propagation effects were of no importance, and the carrier frequency could be absent at all.

All these publications, without precluding fundamental feasibility of the "CPO-based slow light", provide strong evidence that at present there is no necessity to invoke the CPO-based model for interpretation of known experiments on pulse delay in a saturable absorber.

CONCLUSIONS

It should be noted that the discussion about two models of pulse delay in a saturable absorber could arise only for the reason that the authors of [24, 25] missed the fact that the dependences they had found could be comprehensively described by the simplest model of saturable absorber [2, 3, 4] and did not contain any novelty.

One of the authors of the "CPO-based slow light", R.Boyd, in his last review [21] acknowledges that the observed pulse delay can be indeed understood in the sense of the simplest model of saturable absorber of 60's [2], which still, in author's opinion, is equivalent to the "CPO-based slow light".

On the basis of the above consideration, we come to conclusion that all the experimental data on the "CPO-based slow light" completely agree with the simplest model of saturable absorber, have nothing to do with changing group velocity of light, and do not need, for their interpretation, to invoke the effect of coherent population oscillations. Therefore, the term "CPO-based slow light" is, in our opinion, inappropriate in interpretation of the experiments on pulse delay in saturable absorber.

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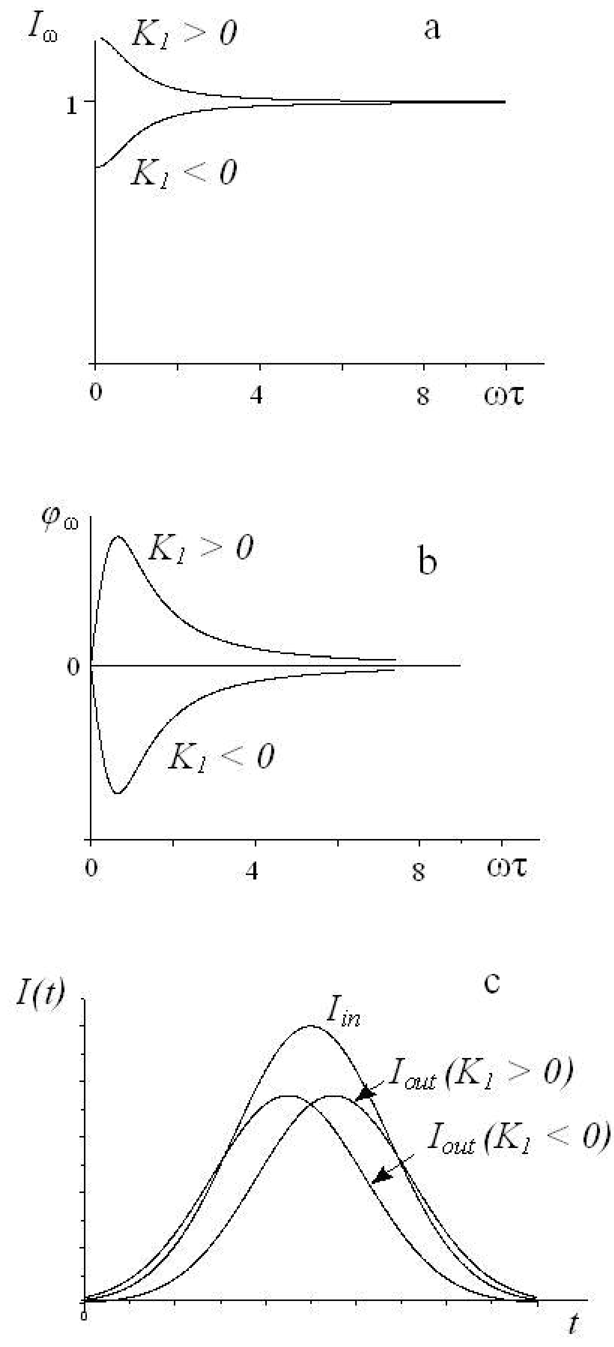

Figure 1: Frequency dependences of the amplitude (a) and phase (b) of the saturable absorber modulation spectrum and distortion of the pulse shape (c).

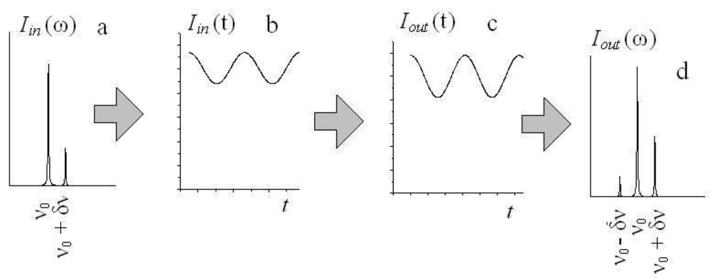

Figure 2: Evolution of optical spectrum of the biharmonic field in the effect of coherent population oscillations. a - optical spectrum of the original field, b - time dependence of the intensity at the entrance of the absorber, c - time dependence of the intensity at the exit of the absorber (increasing modulation depth), d - optical spectrum of the field at the exit.

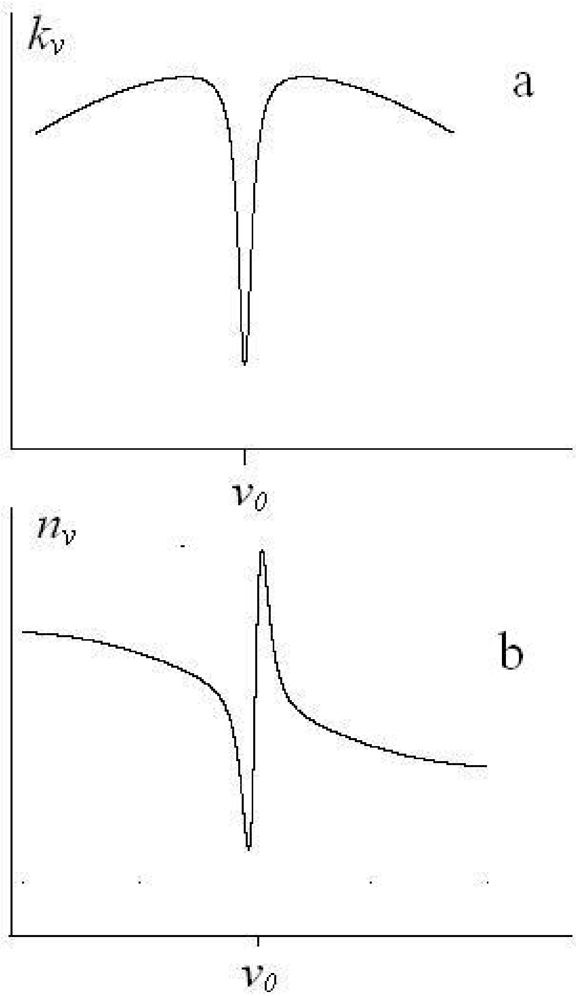

Figure 3: Absorption (a) and refraction (b) spectra of a bleachable absorber detected by the probe light under conditions of CPO (ν_0 is the pump field frequency).